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# System modelling and device development for passive acoustic monitoring of a particulate-liquid process



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## ABSTRACT

This paper presents the development of a passive ultrasonic monitoring system for the detection of acoustic emission (AE) created by chemical particles striking the inner wall of a reactor vessel. The finite element (FE) code PZFlex was used to analyze the complex interactions between chemical particles and the vessel wall. A 4-layer 2D model was developed comprising a liquid load medium and a glass-oil-glass combination corresponding to the jacketed vessel reactor. The model has been experimentally validated with excellent correlation achieved. The excitation function was derived from Hertz's theory and used as the model stimulus corresponding to particles striking the inner glass wall. Analysis of the FE simulations provided the transducer specifications for a passive ultrasonic monitoring system. The system comprises two transducers with complementary characteristics: narrow bandwidth/high sensitivity; wideband/low sensitivity. Importantly, the sensitivity of the resonant transducer provides discrimination of particle concentration. Moreover, the broader bandwidth of the off-resonant device demonstrates potential for *in situ* estimation of particle size. The performance afforded by this approach has considerable potential for real-time process monitoring in the chemicals and pharmaceutical industries.

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## 1. Introduction

Acoustic monitoring techniques offer a significant advantage over optical techniques such as near infrared and Raman spectrometries for process monitoring in that they can be applied to samples that are optically opaque without the need for any sample preparation. In particular, the ability of acoustic waves to penetrate optically opaque media such as stainless steel enables acoustic techniques to be configured to operate in a non-invasive mode of operation, e.g. by sensor attachment or location of a microphone close to the outer vessel wall, without the need for incorporation of a window in the vessel. While there are only a few reports of the use of non-invasive active acoustic techniques for *in situ* process monitoring [1], passive acoustics has been used more widely particularly for the non-invasive monitoring of particulate processes. Passive acoustic monitoring techniques use the acoustic emission (AE) generated by collisions of particles primarily with the inner surface

of vessel or pipe walls [2], although collisions with any internal structures or between particles may also contribute, to determine information related to the status of the process. Measurement of AE has been used successfully to monitor, for example, high-shear granulation [3–6], powder blending [7], drying [8] and various fluidized bed processes [9–15], heterogeneous reactions [16–20], and the transport of powders [21,22], tablets [23] and slurries [24].

A number of experimental and theoretical studies have been published to understand how different factors affect the AE signal generated. In a series of papers, Leach et al. [25–30] investigated AE generated by collisions between spherical, cylindrical and irregular-shaped particles in a rotating vessel. A condenser microphone was used to detect the AE generated by the collisions, with the frequency of emission inversely related to particle size. The same relationship was also observed for the collision of two steel balls [31], and for collisions between glass spheres [32,33] and sediment gravel in water [34]. In addition, it was determined that the amplitude of AE increased with the number of colliding spheres, but that the frequency of AE was unaffected [32]. AE generated by the impact of objects with surfaces has been investigated using a microphone or a transducer attached to the surface of the impacted

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**Table 1**  
Particle information used in both the experimental and simulation investigations.

Mnemonic	Experimental distribution ( $\mu\text{m}$ )	Simulated ( $\mu\text{m}$ )
Size $\alpha$	$0 < x < 251$	200
Size $\beta$	$251 < x < 500$	400
Size $\gamma$	$500 < x < 853$	600

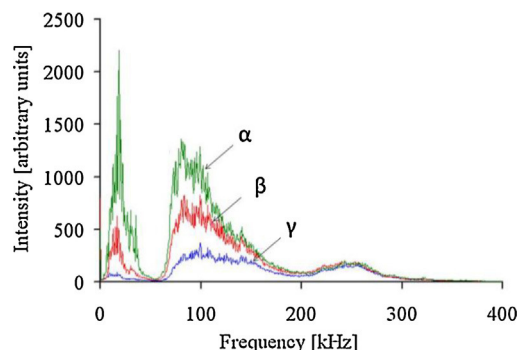
structure. The amplitude of AE arising from collisions between a steel plate and a ball bearing increased as the number of impacting objects was increased, but as for collisions between spheres, the frequency of AE did not change [35]. During the pneumatic conveyance of coal, the maximum frequency of the vibrational modes of the structure excited on impact was found to decrease with an increase in particle size [36,37]. In studies of the mixing of dry powders [7,38] and particles in a liquid [17], the AE signal increased with an increase in the mass and size of particles, while the portion of AE at lower frequencies increased with particle size.

In previous experimental work [16,17], AE generated by the impact of particles in a fluid with the internal wall of a 1 L jacketed glass reactor was studied. More recently, mathematical models were derived to describe the AE generated by particle impacts with the vessel wall [39–41]. The vessel wall was modelled as a single layer circular plate and analytical expressions were derived to describe the impact of particles with the plate. While it was possible to obtain particle size [39,40] and particle concentration [41] information, it becomes increasingly difficult to derive mathematical expressions, which can be solved analytically, describing the propagation of acoustic waves for more complex geometries and material properties. In such cases, it has been shown that a numerical method like finite element (FE) modelling can be used [42,43]. Therefore, in this work, the generation of AE from the impact of particles with a reactor vessel wall for the experimental set-up described in references [16,17] was investigated using FE modelling. A FE model of the experimental set-up comprising a liquid load medium and the glass-oil-glass combination corresponding to the reactor vessel wall structure was developed using PZFlex (Weidlinger Associates Inc., New York, USA). The FE model was used to investigate particle size and concentration characteristics through analysis of the frequency spectra associated with the generated AE from the particle–wall collisions. The results of the investigation have been used to define the ultrasonic transducer system specification for a new passive acoustic monitoring approach, through which a pair of ultrasonic transducers have been designed and fabricated. Interestingly, the complementary characteristics of these transducers can provide additional particulate information from the heterogeneous system under investigation.

## 2. Experimental pilot study

The experimental set-up that was modelled in the present study is described in detail in references [16,17], with the main findings summarized here. The experimental apparatus employed consisted of: a 1 L glass reactor (VWR International, Dorset, UK) with an oil jacket, which was connected to a heater–chiller unit for temperature regulation; a glass stirrer rod and paddle connected to an overhead stirrer motor; a Nano30 AE sensor (Physical Acoustics Limited, Cambridge, UK) attached to the outer wall of the reactor vessel; and a PC for data acquisition and processing.

Broadband AE signals were collected of itaconic acid particles (Sigma Aldrich, Dorset, UK) mixing in 500 mL of toluene (Bamford Laboratories, Rochdale, UK) at 20 °C. To generate different particle size ranges, the itaconic acid was sieved into three fractions (see Table 1). To investigate the effects of particle size and concentration



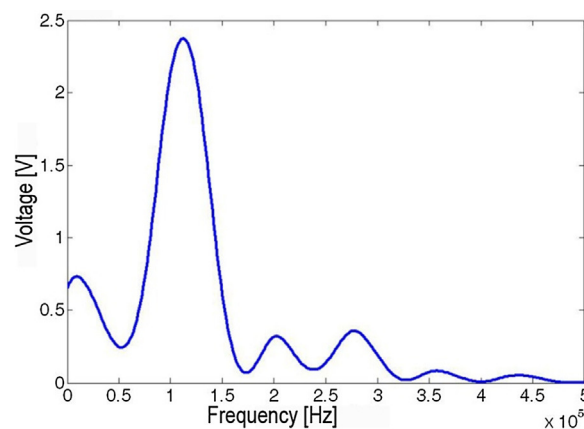
**Fig. 1.** Measured AE power spectra of 40 g dm<sup>−3</sup> of itaconic acid in toluene with particle sizes  $\alpha$ ,  $\beta$  and  $\gamma$ .

on the broadband AE signals, each particle size range of itaconic acid was added stepwise, up to 200 g, to toluene.

Examples of AE spectra of different particle sizes,  $\alpha$ ,  $\beta$  and  $\gamma$ , of 40 g dm<sup>−3</sup> of itaconic acid in toluene are illustrated in Fig. 1. The principal frequencies of interest lay in the 0–350 kHz range, with components above 200 kHz demonstrating less sensitivity to changes in particle sizes to those at lower frequencies.

A variety of data analysis methods were employed to determine both particle size and concentration information. Analyzing the signal area under each AE spectrum, as illustrated in Fig. 1, demonstrated the best opportunity for particle concentration and sizing information. Particle size information was extracted by considering the ratio of signal area between 55 and 200 kHz, where there is clear discrimination between the different particle size ranges, to the overall signal area between 55 and 500 kHz. Full details of the results obtained are given in reference [17].

To characterize the frequency response of different components of the experimental equipment (the jacketed reactor vessel, the Nano30 (Physical Acoustics) transducer and the preamplifier), the inner face of the vessel was excited using a ‘pencil lead break’ (Hsu-Nielsen source) [44] to simulate impulse excitation of the system. Fig. 2 shows the measured frequency spectrum of the system impulse response. On comparing the experimental AE spectra (Fig. 1) with this measured impulse response, the similarity is clearly evident. Therefore, it can be concluded that the acoustic AE signals observed are modified by the frequency transfer function of the transducer, and by the filtering effects of the reactor vessel material and electronic devices.



**Fig. 2.** Experimentally measured spectral profile of the impulse response of the reactor, transducer and preamplifier.

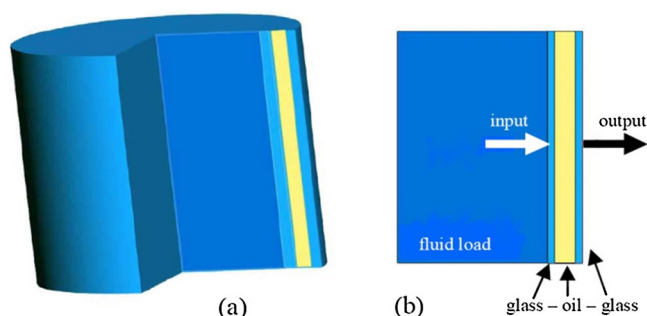


Fig. 3. Finite element model of the vessel reactor.

**Table 2**  
Material properties.

Material	Density ( $\text{kg m}^{-3}$ )	Longitudinal velocity ( $\text{m s}^{-1}$ )	Shear velocity ( $\text{m s}^{-1}$ )
Toluene	860	1225	0
Oil	929	962	0
Glass	2484	6192	3143

### 3. Simulation methodology

#### 3.1. Modelling of the vessel reactor

The approach undertaken to analyze the complex interactions between the chemical particles and the vessel wall used FE modelling techniques based on PZFlex software. Fig. 3(a) shows the three dimensional (3D) 4-layer model developed, comprising a liquid load medium and a glass-oil-glass combination corresponding to the jacketed vessel reactor. Due to computational demand of such a 3D FE model, a 2D longitudinal section of the cylindrical model was utilized throughout this work as illustrated in Fig. 3(b).

Samples of glass and oil from the reactor were collected and their acoustic and physical properties were characterized. These values were obtained by measuring the time of flight in a glass sample of thickness 10 mm and a sample of 50 mL of oil. Table 2 reports the measured acoustic material properties (density, longitudinal velocity, shear velocity) utilized throughout the FE modelling discussed in this paper.

#### 3.2. Simulation of the excitation function

Excitation of the FE model is illustrated in Fig. 3(b) and corresponds to a pressure loading at the inner glass wall using signals derived from the mathematical model of impact of particles of different sizes. The excitation function used as the source of AE was derived from Hertz's theory [40,45] and the model output is the out-of-plane displacement on the outer glass wall in the centre of the model. Assuming an elastic normal impact, each particle colliding against the wall of the vessel generates a force normal to the surface. The determination of the impact source function as a function of time yields the impact time and the magnitude of the force, both of which depend upon particle size and velocity of collision. Table 3 shows the model predictions for contact time and the impact force for particle sizes chosen to correspond

**Table 3**  
Parameters of the predicted particle impact.

Size of particle ( $\mu\text{m}$ )	Experimental distribution	Contact time ( $\mu\text{s}$ )	Force (N)
200	Size $\alpha$	$7.08 \times 10^{-6}$	$4.25 \times 10^{-7}$
400	Size $\beta$	$1.20 \times 10^{-5}$	$9.60 \times 10^{-7}$
600	Size $\gamma$	$1.65 \times 10^{-5}$	$1.50 \times 10^{-6}$

to the particle size distributions utilized in the earlier experimental work, as described in Table 1. Fig. 4 illustrates the temporal and spectral profiles corresponding to particle impact on the wall of the reactor for the three simulated particles sizes described in Table 1. It is important to note that as particle size increases, the impact force amplitude increases and the duration of the contact is longer. Thus, the AE associated with small particles have wider bandwidth when compared to larger particle impacts. It is acknowledged that particle-wall collisions do occur at angles other than the normal direction and moreover, there will be a tangential component involving the particle sliding, or rolling, against the wall. Interestingly, if the particle and wall are sufficiently hard, as in the case considered here, then these tangential components will be second order. Hence, to simplify the excitation scheme for this work only elastic normal impacts are considered.

#### 3.3. FE model validation

Model validation and verification are critical in the development of a simulated model. The mathematical model of the impact of the particle striking a surface has been validated in the literature [46], hence only the validation of the reactor vessel model was performed for this work. It was necessary to reproduce an identical input for the experimental data and predicted model, and then compare the functional output of the two systems. For this purpose, a customized tank was built taking into account the 2D nature of the computerized model. The tank was manufactured using Plexiglass. One wall was composed of two parallel sheets of glass of 3 mm thickness and the 4.5 mm cavity in between contained 500 mL of the same oil that was employed in the reactor heater-chiller. During the validation experiments, a 250 kHz wide-band immersion transducer (Alba Ultrasound Ltd., Glasgow, UK) was positioned in the middle of the vessel and employed as a transmitter. A PVDF hydrophone immersed inside the tank and a PVDF strip (size 30 mm  $\times$  80 mm) attached on the outer wall of the container acted as broadband receivers. An aluminium case was also used to shield the PVDF strip from the environmental noise in the lab. Moreover, the PVDF hydrophone immersed in the tank can be considered acoustically transparent to the propagating ultrasonic energy at the operating frequency of 250 kHz. The receivers were directly connected via a coaxial cable to the Agilent Infiniium oscilloscope which was interfaced to a PC through a GPIB connection to collect received waveforms from the PVDF sensors.

As represented in Fig. 5, the validation of the vessel reactor was conducted in two different stages. In the first stage, a 10 Vpp 20-cycle tone burst, with a centre frequency of 250 kHz was used to drive the ultrasonic transducer and the propagation waveforms were collected by the hydrophone and the PVDF strip receivers.

The first arrival waveform on the hydrophone was selected manually and stored into a file. In the second stage of the validation, the signal output acquired by the hydrophone was employed as the signal input in the FE model as illustrated in Fig. 5(b). In this simple technique, the ultrasonic transducer response can be simulated without knowing its internal characteristic as the signal reproduced is dependent upon the frequency response of the acoustic transmitter. Thus, the predicted output pressure variation was collected at the centre of the simulated model, where the PVDF sensor was theoretically situated and compared with the experimentally measured waveform.

The experimental data and FE model prediction are compared in Fig. 6 to determine if the FE model has sufficient accuracy for use in this simulation programme. The plots show a standing wave pattern established by the layer structure. These standing wave modes arise from interference between the reflected waves and the incident waves. The maximum of the wave is located at 265 kHz and this corresponds to the thickness mode of the ultrasonic transmitter

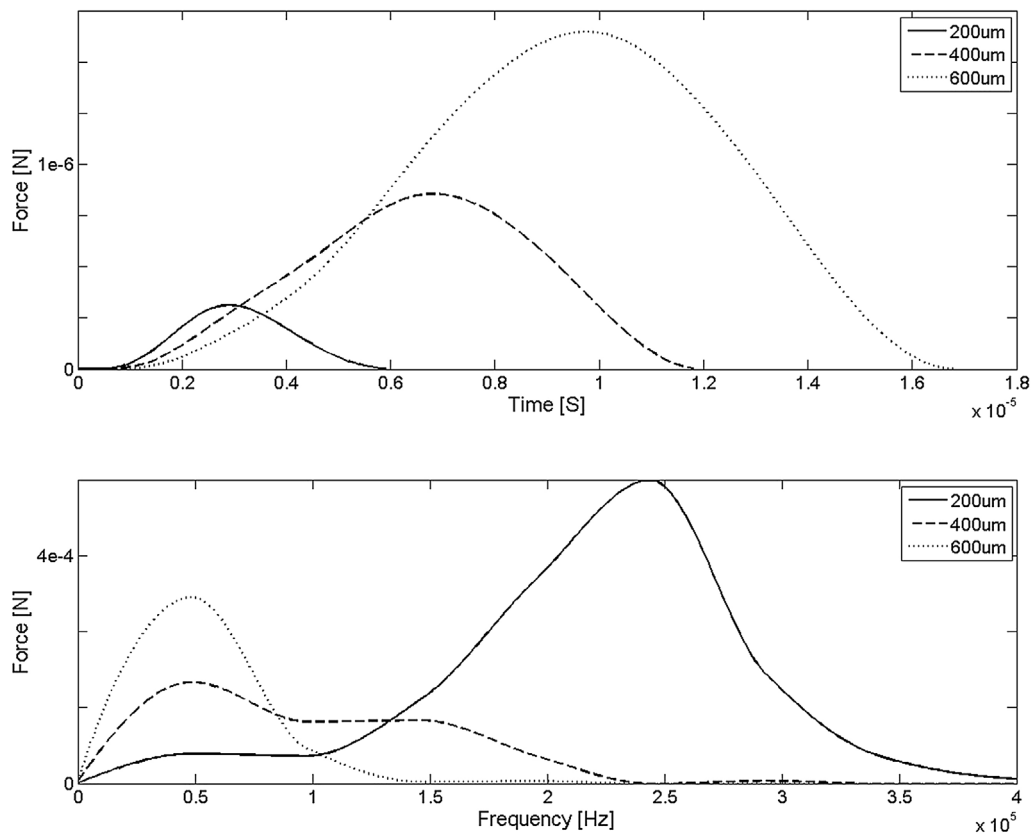


Fig. 4. Predicted impact profiles (temporal and spectral) for 200, 400 and 600  $\mu\text{m}$  particles employed as input excitation function.

used during the validation process. In general, the model shows excellent correlation with the experimental response; the error in the predicted maxima positions is less than 9% for the main four peaks. This good agreement is in part due to the careful characterization of all of the relevant material properties utilized in the model for each layer.

#### 4. FE simulation results

Once the FE model was validated, a number of parameters were selected to provide an insight into the AE generated by the particle impacts on the inner wall of a vessel. One objective of the modelling work was to ascertain if the conceptual model prediction of the system output would be consistent with the experimental feasibility

study and the published theory in the literature regarding particle concentration and particle size effects. The particle sizes used in the simulation study were 200, 400 and 600  $\mu\text{m}$ , which correspond to the experimental particle distribution ranges used in the feasibility study (see Table 1).

##### 4.1. Concentration effects

The effects of different numbers of particles hitting the inner wall of the reactor vessel were simulated. An example of the predicted AE spectra caused by 1, 3 and 7 particles of a fixed particle diameter of 200  $\mu\text{m}$ , impacting in the middle of the inner wall of the vessel is shown in Fig. 7. Here, the multiple excitation scheme is purely theoretical, with the pressure loading function associated

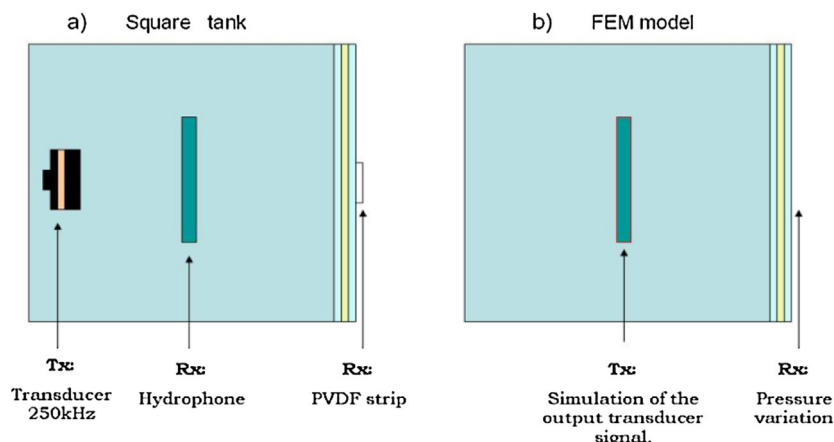


Fig. 5. Illustration of experimental and simulation configurations used in the validation procedure.



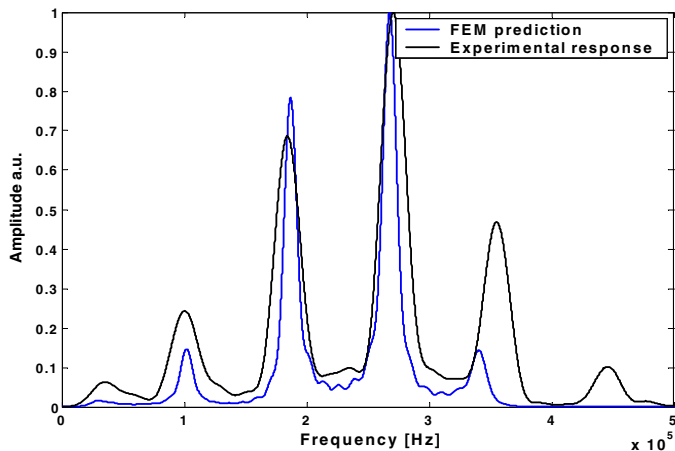


Fig. 6. Comparison of model validation results for experimental response and FEM prediction.

with each particle size applied to the same node in the model to simulate different concentration effects. The pressure variation of the external wall was considered as the output of the system; hence, the shape of the predicted frequency spectra were not influenced by the transfer function of the transducer, as was the case for the experimental results obtained during the feasibility study. Here, only the filtering effects of the reactor structure itself moderate the simulated data. In particular, the reactor vessel behaves as a mechanical low pass filter, with the generated AE restricted to the frequencies below 180 kHz. Interestingly, the original experimental work demonstrates frequency components up to 380 kHz, see Fig. 1 and these higher frequency components correlate directly with resonant modes in the transducer itself. From Fig. 7, the FE model has predicted two resonant modes at 74.4 and 89 kHz corresponding to the resonances associated with the glass-oil-glass configuration, which were calculated to be 75 and 90 kHz, respectively. An increase in the number of impacting particles corresponds to an increase in particle concentration and resulted in an overall increase of AE spectral energy. Importantly, the feasibility study of this heterogeneous reaction produced a similar relationship [17].

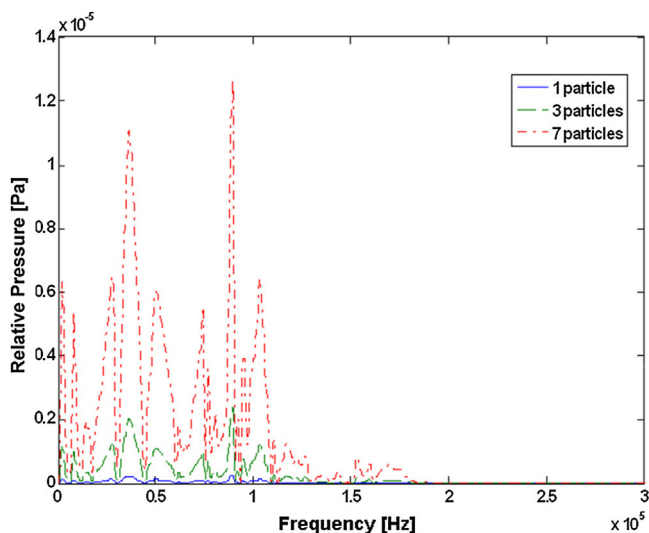


Fig. 7. Predicted AE spectra arising from the impact of 1, 3 and 7 particles with the vessel wall.

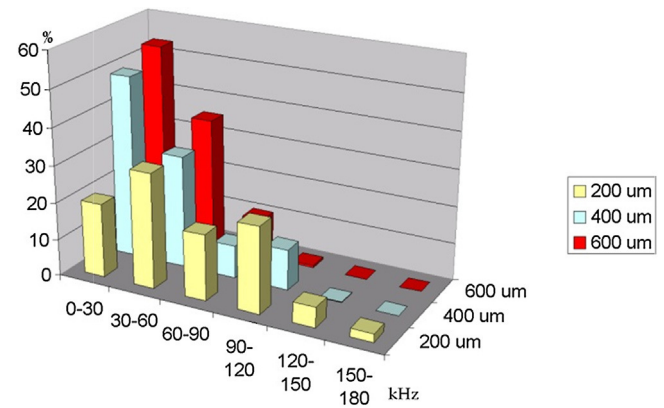


Fig. 8. Histogram of predicted AE spectra arising from the impact of one particle of different sizes.

#### 4.2. Particle size effects

To evaluate the effects of particle size, the excitation function corresponding to one particle of each size detailed in Table 3 was applied as the input to the FE model. The resultant normalized signal profiles are plotted in Fig. 8 and extend between 1 and 170 kHz. Moreover, one thousand particles of fixed size impinging against the first glass layer of the vessel were also simulated to evaluate the effect of multiple particle impacts. In this work, the number of events was limited to 1000 for computational efficiency. To simulate a scenario closer to a practical situation, the simulation software randomized both the spatial impact position and relative times for each AE event associated with the 1000 particle sample. Fig. 9 illustrates the predicted output when the system is subjected to multiple particle impacts. The plot is normalized to highlight the spectra content. A frequency shifting effect is evident, where an increase in the diameter of the object impacting the vessel results in a decrease in AE frequency. Again, this effect is consistent with the experimental work undertaken by the authors and discussed in Section 2 [17].

The changes in signal area between 0 and 150 kHz for the three different particle size ranges as a function of number of particles ( $N$ ) is illustrated in Fig. 10. An increase in the number of impacting particles corresponds to an increase in particle concentration and resulted in an overall increase of AE spectral energy. Importantly, these effects are consistent with published theoretical and experimental studies of collisions between glass spheres [32], a ball

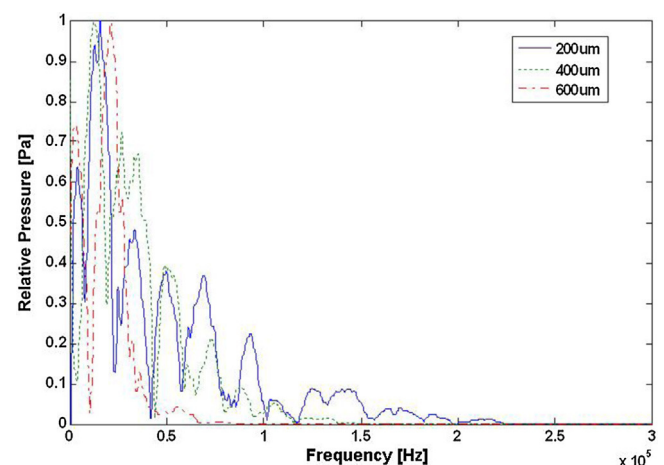


Fig. 9. Predicted AE spectra arising from the randomized impact of 1000 particles of different sizes.

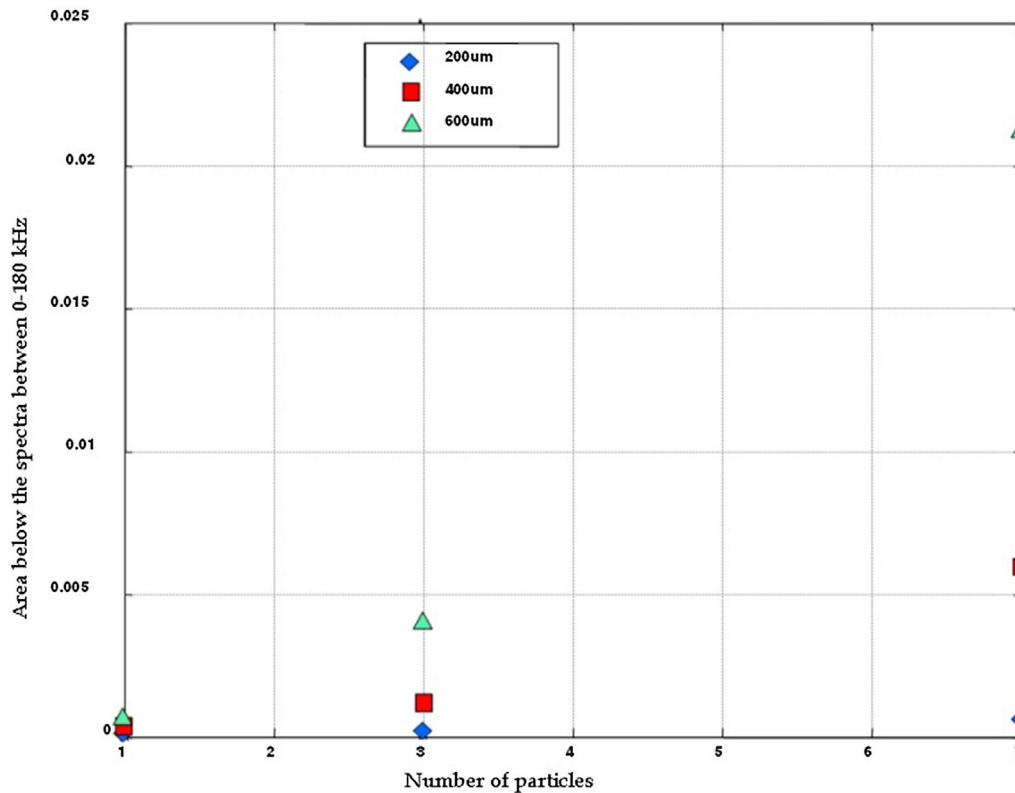


Fig. 10. Relationship between the AE spectral energy and the number of impacting particles and the particle size.

bearing and a plate [35], and other investigations of the mixing of particulate systems [7,16,17,38].

#### 4.3. Discussion of FE simulation results

The FE simulation programme has been used to enhance the understanding of the underlying dynamics of this complex ultrasonic wave propagation system. Importantly, the following statements have to be considered in the context of this work: only a 2D FE model has been utilized; a well-mixed system has been assumed; and the entire system operates at a common temperature. The validated model has provided the following properties associated with particle impacts on a glass reactor vessel:

- The reactor vessel is a highly attenuating system, with only 1.1% of the energy in the horizontal plane transferred through to the reception sensor position.
- The predicted system sensitivity to particle impact at the furthest excitation point in the model is only 3% weaker than an impact in the centre of the model, thus the AE sensor position is not the most critical parameter associated with system SNR. Interestingly, this supports the experimental results acquired during the pilot study and presented in reference [17].
- Particle size has a direct correlation to the energy in the system.
- The AE spectral profiles are strongly dependent on particle size.
- The AE spectral responses are within a 0–180 kHz frequency band, with the main contributions between 10 and 60 kHz and a peak around 40 kHz.

#### 4.4. Ultrasonic transducer specification

Analysis of the simulation results provided the basis of ultrasonic transducer design specifications for use in a passive

monitoring system that provides enhanced sensitivity and operates over the broad frequency range of 0–180 kHz.

It was decided that the most appropriate way to match the ultrasonic system performance with the significant findings of the FE simulation programme was to develop a dual-transducer approach. To enhance the sensitivity of the monitoring system, a resonant 40 kHz ultrasonic transducer was proposed. Whereas, the broadband system requirements could be matched using an off-resonant mode of operation. It was considered that these two transducers

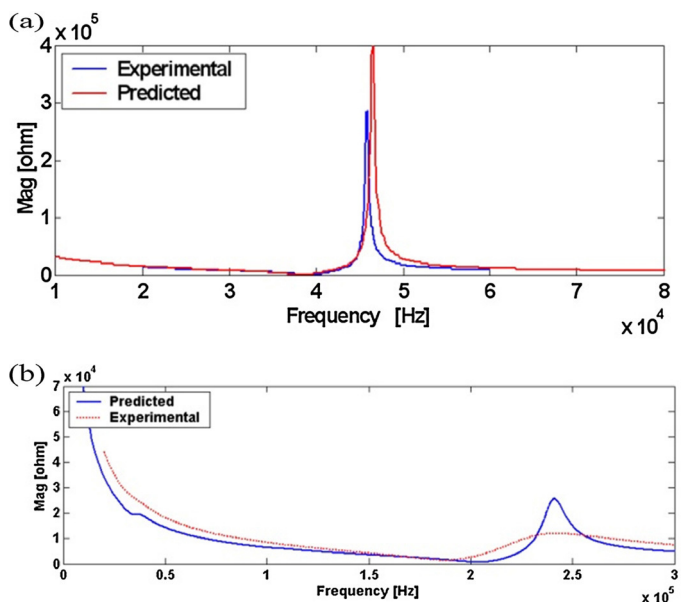
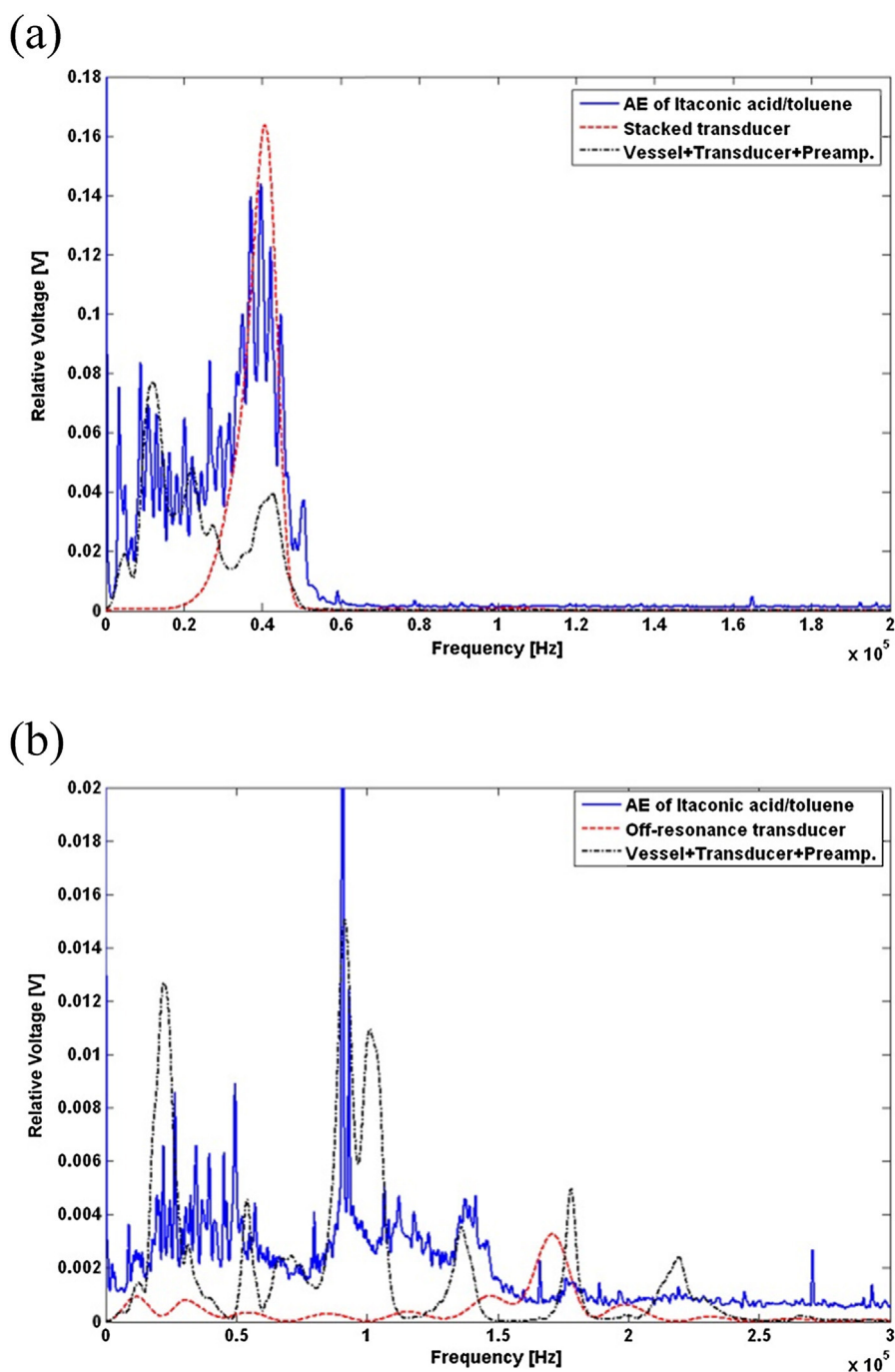


Fig. 11. Magnitude of electrical impedance for (a) stacked device and (b) off-resonance device.



**Fig. 12.** AE spectra of itaconic acid in toluene, and the impulse responses of the transducer and system acquired using (a) the stacked transducer and (b) the off-resonance transducer.

offered complementarity in their monitoring capabilities and could be used to provide both particle concentration and particle size information on heterogeneous systems found in the chemicals and pharmaceutical industries.

## 5. Experimental results

### 5.1. Transducer fabrication

The two ultrasonic transducer designs utilized piezoelectric ceramic composite configurations [47] to tailor the piezoelectric

properties to match the desired specification. In each device, the load medium was the glass wall of the reactor.

The resonant 40 kHz transducer design (stacked device) utilized a stacked 3–1 connectivity composite transducer comprising five layers of PZT4D ceramic connected mechanically in series and electrically in parallel, with a single slot in each vertical side incorporating a soft polymer CIBA/GEIGY CY208/HY956 [48]. The device had an active lateral dimension of 9 mm × 9 mm, encapsulated within a polymer cylinder of diameter 45 mm and the transducer had an overall thickness of 37 mm.

To accommodate the broadband ultrasonic receiver design specification, a 1–3 connectivity piezoelectric ceramic composite



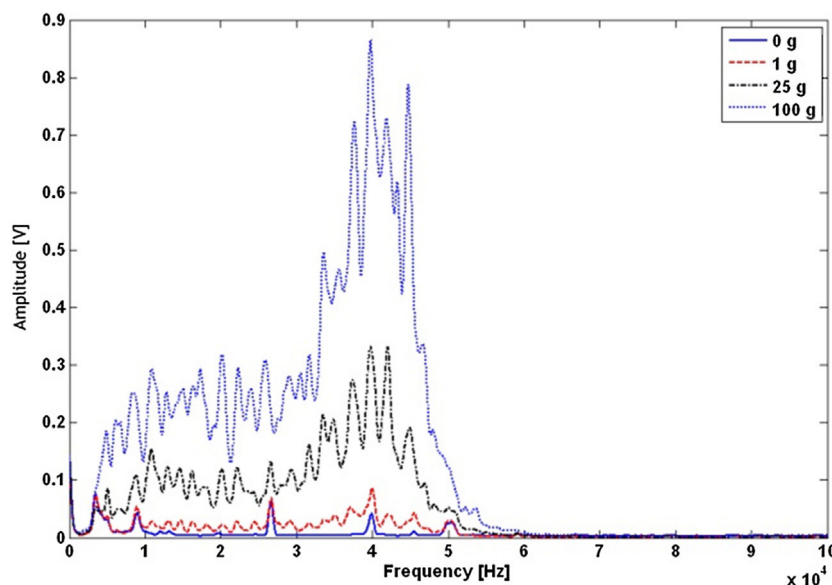


Fig. 13. Power spectra of different masses of itaconic acid in 500 mL of toluene measured using the stacked transducer.

transducer [47,49–51] was designed to operate below its fundamental resonant mode (off-resonance device). Again, PZT4D ceramic was the active phase and the same soft polymer was used as the passive phase in a 30% ceramic volume fraction device with dimensions of 18 mm × 18 mm × 7.8 mm. The fundamental thickness mechanical resonance frequency was designed to occur at 240 kHz, with no other significant resonances below the thickness mode due to the high lateral attenuation of the soft polymer incorporated into the piezocomposite design.

Fig. 11 illustrates the magnitude of the electrical impedance, both experimental and simulated, for each transducer configuration. There is excellent correlation between the theory and experimental data for the stacked configuration, as shown in Fig. 11(a). The agreement is not as good for the off-resonance device, Fig. 11(b), although it is clear that there is only one significant resonance mode below 200 kHz.

Moreover, a pre-amplifier was integrated into each transducer housing to improve system SNR. The pre-amplifier had a voltage gain of 40 dB, with low noise components chosen and pass-band filters to complement the transducer operating characteristics, included in the design.

To evaluate the transducer performance, three experimental measurements were acquired for each configuration: the impulse response of the unloaded transducer; the impulse response of the transducer attached to the vessel and the AE detected by each device from particulate interaction in the vessel. The Hsu-Neilson pencil lead test was used as a wideband excitation source for both of the impulse measurements. The two measurements were then compared with the AE spectra acquired from 100 g of itaconic acid (size  $\beta$ ) suspended in 500 mL of toluene and agitated at a stir rate of 250 rpm. Fig. 12(a) and (b) illustrates the spectral characteristics for the stacked and off-resonant transducer configurations, respectively. It is clearly evident that the stacked transducer provides superior sensitivity and the off-resonant composite operates over a wider frequency range. It should be noted that the frequency spectrum of the off-resonant device is not flat between 0 and 180 kHz. First, the resonant mode of the manufactured device is slightly lower at 170 kHz and there are some lateral mode activity present at 20 and 35 kHz. Nevertheless, this device is considered appropriate for this application as indicated by the wideband response associated with the AE from the particulate suspension shown in Fig. 12(b). It is also interesting to note that the ‘system’ impulse

response, i.e. for the transducer, vessel and pre-amplifier, is dominated by the vessel resonances as observed in the original feasibility study [17].

## 5.2. Concentration effects

Fig. 13 shows the spectra corresponding to different concentrations of itaconic acid of a fixed particle size  $\alpha$  in 500 mL of toluene and employing the stacked transducer as the AE sensor. As expected the peak sensitivity is at the resonance frequency of the device, with discrimination between each particle concentration clearly observed.

The difference in signal energy for the particle size ranges described in Table 1, as a function of concentration, employing the stacked transducer is illustrated in Fig. 14. Assuming AE arising from  $N$  particles impacting with constant velocity on the inner layer of the glass reactor, the intensity or energy will be proportional to the square root of  $N$  [35]. However, power spectra were calculated in this work, so the intensity of power spectra should vary linearly with the mass concentration of particles and hence  $N$ . The variation of energy is approximately linear with concentration

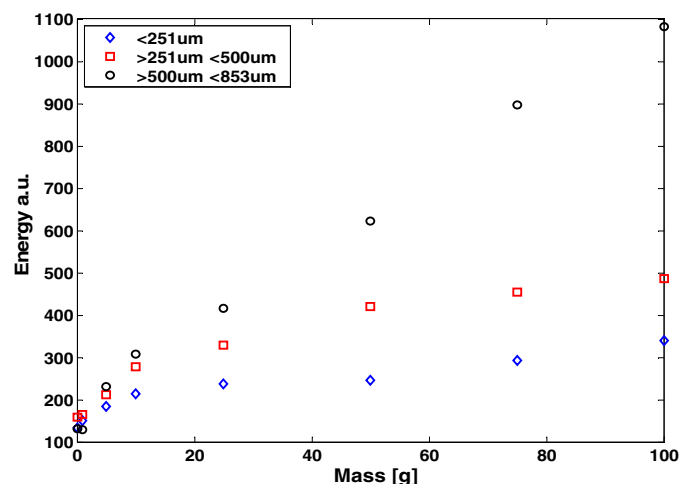


Fig. 14. Effects of both concentration and particle size on the AE signal energy measured using the stacked transducer.

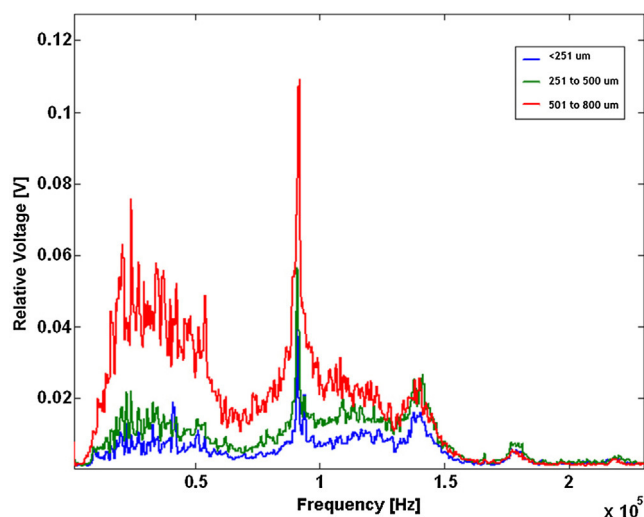


Fig. 15. AE power spectra of different sizes of itaconic acid particle measured using the off-resonant transducer.

up to  $20 \text{ g dm}^{-3}$  (i.e. 10 g in 500 mL) Above this concentration the sensitivity changes due to the saturation of particles in toluene or the presence of other sources of AE (particle–impeller collisions, particle–particles collisions) [17]. In fact, at values above  $20 \text{ g dm}^{-3}$  the degree of non-linearity depends on the particle size, with the largest particles giving a more linear response. From Fig. 14 alone there is no improvement on the information that is achieved using RMS signals because it would not be possible to distinguish between, for example, the signal energy of a high concentration of small particles from a low concentration of large particles.

### 5.3. Particle size effects

Interesting spectral features were apparent when employing the off-resonant transducer. Fig. 15 illustrates the spectra for 100 g of different particle sizes of itaconic acid in 500 mL of toluene collected using the off-resonant transducer. Note that the peak at 90 kHz corresponds to the resonance of the oil layer. The increase in the relative intensity of the signals in the lower frequency regions as particle size increased is in agreement with previously reported studies of the impact of objects with surfaces [7,17,35–38]. An increase in particle size causes an increase in AE magnitude and a decrease in AE frequency owing to a dependence on the particle surface area and impact time, respectively [33]. The frequency

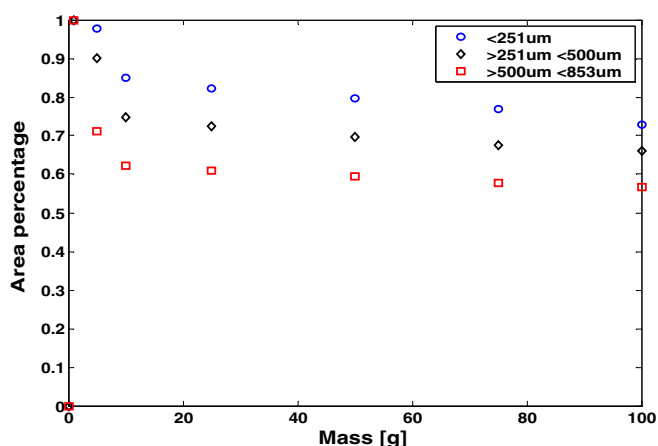


Fig. 16. Percent area of AE spectra, measured using the off-resonant transducer, for different concentrations and particle sizes of itaconic acid in toluene.

shifting effects are not immediately obvious from Fig. 15 and hence, the spectra were dissected into a number of frequency ranges to examine their potential to discriminate between different particle sizes.

The energy of each spectrum in the range between 60 and 200 kHz was computed as a percentage of energy over the total bandwidth (0–200 kHz). Fig. 16 illustrates this approach for different particle sizes; the percentage of area was invariant with concentration, but increased with the particle size range and could be useful for estimating the mean size of particles in liquid. Most commercial AE monitoring systems would miss this type of information as they often convert the signal to a DC level. Importantly, the shift towards lower frequencies, corresponding to an increase in particle size, could be an important index for *in situ* particle size characterization.

## 6. Conclusions

A model-based approach has been described through which the acoustic emission characteristics associated with particle–wall impacts in a reactor vessel have been analyzed to provide a transducer design specification for a non-invasive passive acoustic monitoring system. The transducer specification lends itself to implementation through two separate devices: a low-frequency (40 kHz) sensitive configuration and a broadband (10–180 kHz) off-resonant structure. The complementarity of these two devices has been shown to provide discrimination of particle concentration combined with particle size qualification.

The ability to monitor changes in particle size and concentration information is of great interest to many pharmaceutical and food industry processes. There is typically a compromise between the sensing capabilities of such a monitoring system, with the following key priorities desired: non-invasive; low power; high sensitivity; and particle size discrimination. The dual transducer approach described here offers a route to *in situ* process monitoring by combining all these desirable attributes using ultrasonic transducers. Importantly, it would be possible to integrate both transducer configurations described in this paper into a single package, with adequate lateral damping between the two phases of the transducer vital to minimize crosstalk.

This paper has investigated particle concentration and size and not addressed the practical issues of mixtures with different particle material properties. For example, the AE spectral characteristics for two particles of the same size, but different density, illustrate that the frequency content is similar, with some of the higher frequency components reduced in amplitude for the denser particle case. This would be an additional level of complexity of using the acoustic technique described in this paper. However, it is the authors' proposition that if the mixtures contained different materials, this is where additional techniques such as Raman spectrometry would be needed to provide chemical information and hence, complement the information provided by AE. If a process contained mixtures of different materials, while it would be difficult to deduce the particle size of each material from the AE signal, AE could be used to provide a signature of the process and hence, could be used to detect any process changes.

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